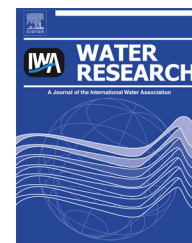


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Experimental and computational investigations of the interactions between model organic compounds and subsequent membrane fouling

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ABSTRACT

The formation of aggregates of sodium alginate and bovine serum albumin (BSA) (as representative biopolymers) with humic acid were detected by Liquid Chromatography (LC) UV₂₅₄ response in the biopolymer region for mixture solutions. BSA interaction with humic acid showed that aggregation occurred both in the presence and absence of calcium, suggesting that multivalent ions did not play a part in the aggregation process. Similar analyses of the alginate interaction with humic acid also showed a positive interaction, but only in the presence of calcium ions. The fouling characteristics for the BSA–humic acid mixture appeared to be significantly greater than the fouling characteristics of the individual solutions, while for the sodium alginate–humic acid mixture, the fouling rate was similar to that of the sodium alginate alone. The effectiveness of hydraulic backwashing, 10–15% reversibility, was observed for the BSA–humic acid mixture, while the % reversibility was 20–40% for the sodium alginate–humic acid mixture. Increased humic acid and DOC rejection were observed for both BSA–humic acid and sodium alginate–humic acid solutions compared to the individual solutions, indicating that the biopolymer filter cakes were able to retain humic acids. When compared with BSA–humic acid mixture solution, greater removal of humic acid was observed for alginate–humic mixture, suggesting that sodium alginate may have a greater capacity for associations with humic acid when in the presence of calcium than BSA. Complementary molecular dynamics simulations were designed to provide insights into the specific mechanisms of interaction between BSA and humic acid, as well as between alginate and humic acid. For the BSA–humic acid system; electrostatic, hydrophobic and hydrogen bonding were the dominant types of interactions predicted, whilst divalent ion-mediated bonding was not identified in the simulations, which supported the LC-results. Similarly for the alginate–humic acid system, the interactions predicted were divalent ion-mediated interactions only and this was also supported the LC results. This work suggests that LC-UV₂₅₄ might be used to identify aggregated biopolymers, and that combined with current characterisation techniques, be used to better explain performance variations between water sources.

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1. Introduction

Membrane filtration in drinking water treatment and wastewater recovery/reuse involves fouling caused by organic matter (Lee et al., 2004). Many studies of organic fouling have focused on one model NOM foulant for the purpose of understanding the fouling mechanism (Schaefer et al., 2000; Lee and Elimelech, 2006). Fouling studies using natural surface waters reported that hydrophilic (non-humic) components of NOM were more significant foulants (Carroll et al., 2000; Gray et al., 2007) than the hydrophobic fraction of NOM (Jucker and Clark, 1994), which consists mainly of organic acids and neutrals.

However, in recent times, the focus of studies on organic fouling has shifted from the study of single model foulants to mixtures. The interaction between organic compounds has been identified as an important mechanism in membrane fouling (Jermann et al., 2007; Gray et al., 2008; Gray et al., 2011; Henderson et al., 2011). Jermann et al. (2007) investigated the effect of molecular interactions within and between humics and polysaccharide on UF fouling mechanisms at organic concentration levels relevant for Swiss lakes. A similar study was also conducted by Katsoufidou et al. (2010). Their studies highlighted that when a mixture of two or more fouling species was present in water, interplay between organic foulants (foulant–foulant) as well as foulant–membrane interaction were observed.

Relatively few studies have reported interactions between different organic compounds with respect to fouling by real waters. For example, Gray et al. (2008), in a study of two different surface waters for different membranes, have described the effect of smaller organic acids and proteins on increasing the fouling rate of high molecular weight compounds. Additionally, Kim et al. (2007) have shown that the fouling potential of filtered waters could be similar to that of the original feed water due to the agglomeration of small molecular weight organic compounds that are present in filtered waters. Clearly, the mechanism of organic fouling of low molecular weight compounds is complex. It may involve a range of organic compounds, the predominant foulant may vary with the source of the water and it may be dependent upon the interactions between various components. Therefore, characterization of such organic interactions, including at the molecular level is important for an understanding of membrane fouling and other water treatment processes, such as organic removal via coagulation or ion exchange.

The aim of this study was to identify possible interactions between organic compounds that are commonly found in natural waters (natural organic matter, NOM) and wastewaters (effluent organic matter, EfOM). Model organic compounds were chosen with structures similar to, or representative of, those considered important in membrane fouling, as well as mixtures of these compounds. More specifically, the compounds used were humic acid, Bovine Serum Albumin (BSA) as an example of a protein and sodium alginate to mimic polysaccharides. Characterizations were performed for solutions in a synthetic background electrolyte of similar composition to that of a municipal wastewater. Liquid chromatography with organic carbon detection (LC-UV₂₅₄-OCD)

and LC coupled with a photo-diode array (PDA) detector were used to probe for interactions in the waters themselves. The ability to identify the presence of interactions between various compounds was thus assessed, and complementary molecular dynamics simulations were used to predict the range of specific interactions that may occur at the molecular level. The computational results were then reconciled with the experimental data. In addition, the fouling response of the specific mixtures with a hydrophobic polypropylene membrane was explored, based on the experimental and theoretical findings observed with the presence of interactions between various organic compounds.

2. Materials and methods

2.1. Organic foulants

Sodium alginate from brown algae (Sigma-Aldrich), bovine serum albumin (BSA, Sigma-Aldrich), and humic acid (HA) (Fluka) were selected as model organic foulants to represent polysaccharides, proteins, and humic acid found in EfOM. Both sodium alginate and BSA (biopolymers) were chosen to represent the high molecular weight compounds present in surface waters and wastewaters, whereas the smaller humic acid was selected to represent the hydrophobic characteristics of organic matter.

2.1.1. Feed solution preparation

Stock solutions (1 g L⁻¹) were prepared by dissolving each of the foulants in deionized (MilliQ) water. Stock solution of humic acid was adjusted to pH 10 using 5 M sodium hydroxide (NaOH) solution to ensure complete dissolution of the foulants. While raising the pH improved dissolution of the humic acid, a small UV₂₅₄ biopolymer peak was still detected for humic acid solution indicating some residual agglomeration within the humic acid solution.

Model foulant solutions for organic characterization experiments were prepared from the stock solution. 100 ml of each model foulant solution was prepared by diluting the required amount of each foulant stock solution to typically 25 mg L⁻¹. In order to investigate the interactions between specific species, a range of mixtures containing one biopolymer (either BSA or alginate) and humic acid were prepared and analysed via LC-PDA. When determining the extent of interaction between BSA or alginate and humic acid via UV₂₅₄ in the biopolymer region, the residual humic acid peak in this region was subtracted from the peak for the mixture solutions.

The ionic environment for experiments in electrolyte solution consisted of NaCl (0.003 M), CaCl₂ (0.001 M), KCl (0.0004 M) and MgCl₂ (0.0004 M) prepared in deionised water. The solutions were adjusted to pH 7–7.5 with 0.01 M hydrochloric acid. The total ionic strength (circa $I = 0.77 \times 10^{-2}$ M, 420 mg L⁻¹) was confirmed by conductivity measurements. The prepared pH, ionic strength and cation concentrations were chosen because of their similarity to a local secondary effluent wastewater. Table 1 summarises the foulant solutions prepared for organic characterization.

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